

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner  
 US Department of Commerce  
 United States Patent and Trademark  
 Office, PCT  
 2011 South Clark Place Room  
 CP2/5C24  
 Arlington, VA 22202  
 ETATS-UNIS D'AMERIQUE  
 in its capacity as elected Office

<b>Date of mailing</b> (day/month/year) 16 November 2000 (16.11.00)	
<b>International application No.</b> PCT/EP00/02878	<b>Applicant's or agent's file reference</b> F-748 WO
<b>International filing date</b> (day/month/year) 28 March 2000 (28.03.00)	<b>Priority date</b> (day/month/year) 30 March 1999 (30.03.99)
<b>Applicant</b> DEMAIN, Axel	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
 26 October 2000 (26.10.00)

☐ in a notice effecting later election filed with the International Bureau on:  
 \_\_\_\_\_

2. The election ☒ was

☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer S. Mafla
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

## PATENT COOPERATION TREATY

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From the INTERNATIONAL BUREAU

NOTIFICATION OF THE RECORDING  
OF A CHANGE(PCT Rule 92bis.1 and  
Administrative Instructions, Section 422)

To:

ATOFINA RESEARCH  
Patent Dept.  
Zone Industrielle C  
B-7181 Seneffe  
BELGIQUE

<b>Date of mailing</b> (day/month/year) 26 October 2001 (26.10.01)	
<b>Applicant's or agent's file reference</b> F-748 WO	<b>IMPORTANT NOTIFICATION</b>
<b>International application No.</b> PCT/EP00/02878	<b>International filing date</b> (day/month/year) 28 March 2000 (28.03.00)

1. The following indications appeared on record concerning:		
<input checked="" type="checkbox"/> the applicant	<input type="checkbox"/> the inventor	<input type="checkbox"/> the agent <input type="checkbox"/> the common representative
Name and Address FINA RESEARCH S.A. Zone Industrielle C B-7181 Seneffe Belgium	State of Nationality BE	State of Residence BE
	Telephone No.	
	Facsimile No.	
	Teleprinter No.	
2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:		
<input type="checkbox"/> the person	<input checked="" type="checkbox"/> the name	<input type="checkbox"/> the address <input type="checkbox"/> the nationality <input type="checkbox"/> the residence
Name and Address ATOFINA RESEARCH Zone Industrielle C B-7181 Seneffe Belgium	State of Nationality BE	State of Residence BE
	Telephone No.	
	Facsimile No.	
	Teleprinter No.	
3. Further observations, if necessary:		
4. A copy of this notification has been sent to:		
<input checked="" type="checkbox"/> the receiving Office	<input type="checkbox"/> the designated Offices concerned	
<input type="checkbox"/> the International Searching Authority	<input checked="" type="checkbox"/> the elected Offices concerned	
<input type="checkbox"/> the International Preliminary Examining Authority	<input type="checkbox"/> other:	

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland  Facsimile No.: (41-22) 740.14.35	Authorized officer  S. Buttay  Telephone No.: (41-22) 338.83.38
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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>F-748 WO</b>	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/EP00/02878</b>	International filing date (day/month/year) <b>28/03/2000</b>	Priority date (day/month/year) <b>30/03/1999</b>
International Patent Classification (IPC) or national classification and IPC <b>D01F6/46</b>		
Applicant <b>FINA RESEARCH S.A. et al.</b>		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 4 sheets, including this cover sheet.

- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand <b>26/10/2000</b>	Date of completion of this report <b>08.12.2000</b>
Name and mailing address of the international preliminary examining authority:  <b>European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465</b>	Authorized officer  <b>Adams, F</b>  Telephone No. +49 89 2399 8511 

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP00/02878

## I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).*):

### Description, pages:

1-10 as originally filed

### Claims, No.:

1-19 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP00/02878

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes:	Claims	1-19
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-19
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-19
	No:	Claims	

2. Citations and explanations  
**see separate sheet**

Ad V:

- 1). The present application satisfies the criteria set forth in Article 33(2) and 33(3) PCT because the subject-matter of the claims is new and inventive in respect of prior art as defined in the regulations (Rule 64(1)-(3) PCT).

None of the cited documents discloses a polypropylene blend including from 0.3 - 50 wt-% of syndiotactic polypropylene having a multimodal molecular weight distribution, and at least 50 wt-% of an isotactic polypropylene.

None of the cited documents deals with the problem of improving processability of polypropylene blends which allows higher spinning speeds when preparing fibers.

- 2). US-A-5648428 (D1, cited in the application) discloses a process for producing a blend of syndiotactic polypropylene and isotactic polypropylene in a single reactor wherein the blend has a broad molecular weight distribution (MWD=13). The amount of syndiotactic polypropylene in the blend is greater than 10 %. Nothing is said about a blend wherein the syndiotactic polypropylene itself has a multimodal molecular weight distribution. Nothing is said about the problem of improving the spinning speed when preparing fibers.

EP-A-0634505 (D2, cited in the application) discloses blends of syndiotactic polypropylene with a narrow molecular weight distribution and isotactic polypropylene. Nothing is said about multimodal syndiotactic polypropylene. Nothing is said about the problem of improving the spinning speed when preparing fibers.

EP-A-0789096 (D3, cited in the application) discloses blends of syndiotactic polypropylene and isotactic polypropylene. Nothing is said about multimodal syndiotactic polypropylene. Nothing is said about the problem of improving the spinning speed when preparing fibers.

# PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>F-748 WO</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/EP 00/ 02878</b>	International filing date (day/month/year) <b>28/03/2000</b>	(Earliest) Priority Date (day/month/year) <b>30/03/1999</b>
Applicant <b>FINA RESEARCH S.A. et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 03 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

### 1. Basis of the report

a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of Invention is lacking** (see Box II).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☒ None of the figures.

27 JUN 2001

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

14

Applicant's or agent's file reference F-749 WO	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP00/02879	International filing date (day/month/year) 28/03/2000	Priority date (day/month/year) 30/03/1999
International Patent Classification (IPC) or national classification and IPC C08L23/06		
Applicant FINA RESEARCH S.A. et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 4 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 4 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand  26/10/2000	Date of completion of this report  25.06.2001
Name and mailing address of the international preliminary examining authority:   European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Knutzen-Mies, K  Telephone No. +49 89 2399 8525  



**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP00/02879

**i. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17):*

**Description, pages:**

1-17 as originally filed

**Claims, No.:**

1-29 with telefax of 07/03/2001

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☒ the claims, Nos.: 30,31
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP00/02879

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes:	Claims	1-29
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-29
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-29
	No:	Claims	

2. Citations and explanations  
**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP00/02879

**ad section V.:**

None of the documents cited in the international search report discloses or fairly suggests a multimodal polyethylene having a density between 0.930 and 0.955 g/cm<sup>3</sup> and comprising a high molecular weight fraction having a density of 0.930 g/cm<sup>3</sup> or less and a high load melt index of 0.30 g/10 min or less.

The subject matter of claims 1 - 29 of the present application therefore satisfies the requirements of Article 33(2) -(4) PCT.

## CLAIMS:

1. A multimodal polyethylene, having a density of from 0.930 to 0.955 g/cm<sup>3</sup> and a shear ratio (SR) of 18 or more and comprising at least 20 % by weight of a high molecular weight fraction, which high molecular weight fraction has:
  - (a) a density ( $\rho$ ) of 0.930 g/cm<sup>3</sup> or less; and
  - (b) a high load melt index (HLMI) of 0.30 g/10 mins or less.
2. A multimodal polyethylene according to claim 1, wherein the density of the high molecular weight fraction is 0.925 g/cm<sup>3</sup> or less.
3. A multimodal polyethylene according to claim 2, wherein the density of the high molecular weight fraction is 0.923 g/cm<sup>3</sup> or less.
4. A multimodal polyethylene according to any preceding claim, wherein the HLMI of the high molecular weight fraction is 0.20 g/10 mins or less.
5. A multimodal polyethylene according to any preceding claim, comprising 45% by weight or more of the high molecular weight fraction.
6. A multimodal polyethylene according to any preceding claim, having a final HLMI of 30g/10 mins or less.
7. A multimodal polyethylene according to claim 6, having a final HLMI of 5-10 g/10 mins or less.
8. A multimodal polyethylene according to any preceding claim, which polyethylene has a value of 2000 hours or more in a full notch creep test.
9. A multimodal polyethylene according to claim 8, which polyethylene has a value of 5000 hours or more in a full notch creep test.

10. A multimodal polyethylene according to any preceding claim, which polyethylene has a value of 5000 hours or more in a notch pipe test.
11. A multimodal polyethylene according to claim 10, which polyethylene has a value of 10000 hours or more in a notch pipe test.
12. A multimodal polyethylene according to any preceding claim, which is a bimodal or a trimodal polyethylene.
13. A multimodal polyethylene according to any preceding claim, which is a PE 100 or a PE 80 polyethylene.
14. A multimodal polyethylene according to any preceding claim, wherein the high molecular weight fraction is physically blended with the remaining fractions or is chemically blended with the remaining fractions.
15. A method for the production of a multimodal polyethylene as defined in any preceding claim, which method comprises mixing a high molecular weight polyethylene fraction as defined in any of claims 1 to 5 with one or more lower molecular weight fractions.
16. A method according to claim 15, wherein the high molecular weight fraction is produced separately from one or more lower molecular weight fractions, and said high and lower molecular weight fractions are mixed together in a physical blending process.
17. A method according to claim 15, wherein the high molecular weight fraction is produced in the presence of one or more lower molecular weight fractions, or one or more lower molecular weight fractions are produced in the presence of the high molecular weight fraction, such that said high and lower molecular weight fractions are mixed together in a chemical blending process.
18. A method according to any of claims 15 to 17, wherein the high molecular weight fraction is produced by polymerising ethylene in the presence of an alpha-olefin co-monomer having from 3-10 carbon atoms.

19. A method according to claim 18, wherein the co-monomer is butene or hexene.
20. A multimodal polyethylene obtainable according to a method as defined in any of claims 15 to 19.
21. A polyethylene pipe comprising a multimodal polyethylene having a density of from 0.930 to 0.955 g/cm<sup>3</sup> and a high molecular weight fraction, which high molecular weight fraction has a density ( $\rho$ ) and a high load melt index (HLMI), which satisfy the following relationship:

$$\rho \times \text{HLMI} \leq 0.37$$

wherein the units of density are g/cm<sup>3</sup> and the units of HLMI are g/10 mins, and the density ( $\rho$ ) is 0.930 g/cm<sup>3</sup> or less.

22. A pipe according to claim 21, in which the multimodal polyethylene has at least 20 % by weight of the high molecular weight fraction, and the high molecular weight fraction has a high load melt index (HLMI) of 0.40 g/10 mins or less.
23. A pipe according to claim 22, in which the multimodal polyethylene is as defined in any of claims 1 to 16 and 22.
24. A method of pipe installation comprising forming a hole or trench for receiving a pipe and installing a polyethylene pipe as defined in any of claims 21 to 23 in the hole or trench.
25. A method according to claim 24, which method is a no-sand installation method, the pipe being installed in the hole or trench directly in contact with the earth.
26. A method for re-lining a pipe comprising installing a polyethylene pipe as defined in any of claims 21 to 23 in an existing pipe.

27. Use of a multimodal polyethylene having a density of from 0.930 to 0.955 g/cm<sup>3</sup> and comprising a high molecular weight fraction, which high molecular weight fraction has a density ( $\rho$ ) and a high load melt index (HLMI), which satisfy the following relationship:

$$\rho \times \text{HLMI} \leq 0.37$$

wherein the units of density ( $\rho$ ) are g/cm<sup>3</sup> and the units of HLMI are g/10 mins, wherein the high density multimodal polyethylene comprises at least 20 % by weight of the high molecular weight fraction and the density ( $\rho$ ) is 0.930 g/cm<sup>3</sup> or less and the high load melt index (HLMI) is 0.40 g/10 mins or less, in a polyethylene article to provide the article with enhanced creep resistance to stress-cracking as determined by the notch-pipe test (NPT) of EN 33479.

28. Use according to claim 27, in which the multimodal polyethylene is as defined in any of claims 1 to 14 and 22.

29. Use according to claim 27 or claim 28, wherein the article is a polyethylene pipe.

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>7</sup> :</b> <b>D01F 6/46, C08L 23/10</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 00/60149</b> <b>(43) International Publication Date:</b> 12 October 2000 (12.10.00)
<b>(21) International Application Number:</b> PCT/EP00/02878 <b>(22) International Filing Date:</b> 28 March 2000 (28.03.00) <b>(30) Priority Data:</b> 99106462.7 30 March 1999 (30.03.99) EP <b>(71) Applicant (for all designated States except US):</b> FINA RE- SEARCH S.A. [BE/BE]; Zone Industrielle C, B-7181 Sen- effe (BE). <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only):</b> DEMAIN, Axel [BE/BE]; Rue de Sart, 21, B-1457 Tourinnes-Saint-Lambert (BE). <b>(74) Common Representative:</b> FINA RESEARCH S.A.; Patent Dept., Zone Industrielle C, B-7181 Seneffe (BE).		<b>(81) Designated States:</b> AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> POLYPROPYLENE FIBRES		
<b>(57) Abstract</b>  A polypropylene blend including from 0.5 to 50 % by weight of a syndiotactic polypropylene having a multimodal molecular weight distribution and at least 50 % by weight of an isotactic polypropylene.		



*FOR THE PURPOSES OF INFORMATION ONLY*

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

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BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
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CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

POLYPROPYLENE FIBRES

The present invention relates to polypropylene fibres and to fabrics produced from polypropylene fibres.

Polypropylene is well known for the manufacture of fibres, particularly for manufacturing non woven fabrics.

EP-A-0789096 discloses such polypropylene fibres which are made of a blend of syndiotactic polypropylene (sPP) and isotactic polypropylene (iPP). That specification discloses that by blending from 0.3 to 3% by weight of sPP, based on the total polypropylene, to form a blend of iPP-sPP, the fibres have increased natural bulk and smoothness, and non-woven fabrics produced from the fibres have an improved softness. Moreover, that specification discloses that such a blend lowers the thermal bonding temperature of the fibres. Thermal bonding is employed to produce the non-woven fabrics from the polypropylene fibres.

The specification discloses that the isotactic polypropylene comprises a homopolymer formed by the polymerisation of propylene by Ziegler-Natta catalysis.

WO-A-96/23095 discloses a method for providing a non-woven fabric with a wide bonding window in which the non-woven fabric is formed from fibres of a thermoplastic polymer blend including from 0.5 to 25wt% of syndiotactic polypropylene. The syndiotactic polypropylene may be blended with a variety of different polymers, including isotactic polypropylene. The specification includes a number of examples in which various mixtures of syndiotactic polypropylene with isotactic polypropylene were produced. The isotactic polypropylene comprised commercially available isotactic polypropylene, which is produced using a Ziegler-Natta catalyst. It is disclosed in

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the specification that the use of syndiotactic polypropylene widens the window of temperature over which thermal bonding can occur, and lowers the acceptable bonding temperature.

WO-A-96/23095 also discloses the production of fibres from blends including syndiotactic polypropylene which are either bi-component fibres or bi-constituent fibres. Bi-component fibres are fibres which have been produced from at least two polymers extruded from separate extruders and spun together to form one fibre. Bi-constituent fibres are produced from at least two polymers extruded from the same extruder as a blend. Both bi-component and bi-constituent fibres are disclosed as being used to improve the thermal bonding of Ziegler-Natta polypropylene in non-woven fabrics. In particular, a polymer with a lower melting point compared to the Ziegler-Natta isotactic polypropylene, for example polyethylene, random copolymers or terpolymers, is used as the outer part of the bi-component fibre or blended in the Ziegler-Natta polypropylene to form the bi-constituent fibre.

EP-A-0634505 discloses improved propylene polymer yarn and articles made therefrom in which for providing yarn capable of increased shrinkage syndiotactic polypropylene is blended with isotactic polypropylene with there being from 5 to 50 parts per weight of syndiotactic polypropylene. It is disclosed that the yarn has increased resiliency and shrinkage, particularly useful in pile fabric and carpeting. It is disclosed that the polypropylene blends display a lowering of the heat softening temperature and a broadening of the thermal response curve as measured by differential scanning calorimetry as a consequence of the presence of syndiotactic polypropylene.

US-A-5269807 discloses a suture fabricated from syndiotactic polypropylene exhibiting a greater flexibility than a comparable suture manufactured from isotactic polypropylene. The syndiotactic polypropylene may be blended with, *inter alia*,

isotactic polypropylene.

EP-A-0451743 discloses a method for moulding syndiotactic polypropylene in which the syndiotactic polypropylene may be blended with a small amount of a polypropylene having a substantially isotactic structure. It is disclosed that fibres may be formed from the polypropylene. It is also disclosed that the isotactic polypropylene is manufactured by the use of a catalyst comprising titanium trichloride and an organoaluminium compound, or titanium trichloride or titanium tetrachloride supported on magnesium halide and an organoaluminium compound, i.e. a Ziegler-Natta catalyst.

EP-A-0414047 discloses polypropylene fibres formed of blends of syndiotactic and isotactic polypropylene. The blend includes at least 50 parts by weight of the syndiotactic polypropylene and at most 50 parts by weight of the isotactic polypropylene. It is disclosed that the extrudability of the fibres is improved and the fibre stretching conditions are broadened.

US-A-5648428 discloses a process for producing a polymer blend in a single reactor, specifically a polymer blend of isotactic polyolefin and syndiotactic polyolefin which may comprise polypropylene. The single reactor is provided with a catalyst system comprising a combination of at least one metallocene catalyst and at least one conventional supported Ziegler-Natta catalyst.

These known polypropylenes suffer from the disadvantage that the processability of the iPP/sPP blends needs to be improved, particularly for spinning fibres, so that higher spinning speeds can be employed before fibre breakage occurs or the incidence of breakage at any given spinning speed is reduced.

It is further known to produce syndiotactic polypropylene using

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metallocene catalysts as has been disclosed for example in US-A-4794096.

WO-A-96/35729 discloses a process for preparing and using a supported metallocene-alumoxane catalyst which is used for the polymerisation or copolymerisation of olefins to produce an olefin polymer or copolymer having a broad and bimodal molecular weight distribution.

It is an aim of the present invention to produce a blend of iPP and sPP which has improved processability, particularly when forming spun fibres. It is a further aim of the present invention to provide fibres, and fabrics in particular non-woven fabrics made from those fibres, which have been produced from that blend.

The present invention provides a polypropylene blend including from 0.3 to 50% by weight of a syndiotactic polypropylene having a multimodal molecular weight distribution and at least 50% by weight of an isotactic polypropylene.

The sPP is typically bimodal.

The isotactic polypropylene (iPP) may have been produced using a Ziegler-Natta catalyst and may be a homopolymer or copolymer and may have a monomodal or multimodal molecular weight distribution.

Preferably, the syndiotactic polypropylene (sPP) concentration in the sPP/iPP blend is from 0.3 to 15wt%, more preferably from 1 to 10wt%. The fibre may be a two component sPP/iPP blend.

Preferably, the iPP is a homopolymer, copolymer, being either a random or block copolymer, or terpolymer of isotactic polypropylene.

Typically, the isotactic polypropylene has a melting temperature in the range of from 159 to 169°C, more typically from 161 to 165°C. The iPP employed in accordance with the invention has a molecular weight distribution typically having a dispersion index D of from 3.5 to 9, more preferably from 3.5 to 6.5. The dispersion index D is the ratio  $M_w/M_n$ , where  $M_w$  is the weight number average molecular weight and  $M_n$  is the number average molecular weight of the polymer. The iPP typically has a peak in the molecular weight distribution of around 35,000 to 60,000 kDa. The isotactic polypropylene may have an  $M_n$  of from 35,000 to 45,000 kDa.

The isotactic polypropylene preferably has a melt flow index (MFI) of from 1 to 90g/10mins, more preferably from 10 to 60g/10mins. In this specification the MFI values are those determined using the procedure of ISO 1133 using a load of 2.16kg at a temperature of 230°C.

The properties of two typical iPP resins for use in the invention are specified in Table 1.

The multimodal sPP, preferably bimodal sPP, is preferably a homopolymer or a random copolymer with a comonomer content of from 0.1 to 1.5wt%, more preferably from 0.1 to 1wt%. The multimodal sPP may however be a block copolymer with a higher comonomer content or a terpolymer. If the comonomer content is above 1.5wt%, the sPP tends to become sticky, thus resulting in problems when spinning the fibres or thermally bonding the fibres. The comonomer content is selected so as to decrease the melting point of the sPP/iPP blend below 130°C. A lower melting point can also be obtained by using particular catalysts and/or process conditions during polymerisation of the sPP. Preferably, the sPP has a melting temperature of up to about 130°C. The sPP

typically has two melting peaks, one being around 110°C and the other being around 125°C. The sPP typically has an MFI of from 0.1 to 1000g/10min, more typically from 1 to 60g/10min. The multimodal sPP may have an Mn of from 35,000 to 40,000 kDa. The properties of a typical bimodal sPP for use in the invention are specified in Table 1.

The sPP has a slightly narrower molecular weight distribution than for the iPP, wherein typically D may be from 3 to 6, more typically around 4 and has a peak of the molecular weight distribution at around 20,000 to 35,000 kDa. The sPP and the iPP may have substantially similar values for Mn. In view of the closeness between the peaks and the overlap of the molecular weight distributions of the sPP and the iPP, those two components can readily be blended together.

The present invention further provides a fibre produced from the polypropylene blend in accordance with the invention.

The present invention further provides a fabric produced from the polypropylene fibre of the invention.

The present invention yet further provides a product including that fabric, the product being selected from among others a filter, personal wipe, diaper, feminine hygiene product, incontinence product, wound dressing, bandage, surgical gown, surgical drape and protective cover.

The present invention is predicated on the discovery by the present inventor that when blended with iPP, multimodal sPP, preferably bimodal sPP, improves the processability of the polypropylene, particularly to form fibres in a spinning process, as compared to monomodal sPP.

The present inventor has found that when used in an amount of

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only about 2wt% sPP in the sPP/iPP blend, the spinnability is increased when sPP is multimodal, preferably bimodal.

The use of multimodal e.g. bimodal sPP in blends with iPP in accordance with the invention tends to provide fibres which can be more readily spun as compared to known fibres comprised of iPP blended with monomodal sPP. The molecular weight distributions of iPP and the multimodal sPP overlap, and as a result of the provision of sPP having a broad multimodal molecular weight distribution this provides increased overlap between the two distributions as compared to monomodal sPP, thereby improving the blend, and also provides a higher proportion of short chains in the combined molecular weight distribution for the blend. This has been found to result in an increase in the maximum spin speed before breakage for the fibres of the multimodal sPP/iPP blends in accordance with the invention.

The fibres produced in accordance with the invention may be either bi-component fibres or bi-constituent fibres. For bi-component fibres, iPP and sPP are fed into two different extruders. Thereafter the two extrudates are spun together to form single fibres. For the bi-constituent fibres, blends of sPP/iPP are obtained by: dry blending pellets, flakes or fluff of the two polymers before feeding them into a common extruder; or using pellets or flakes of a blend of sPP and iPP which have been extruded together and then re-extruding the blend from a second extruder.

When the blends of multimodal sPP/iPP are used to produce fibres in accordance with the invention, for the production of spunlaid fibres, a typical extrusion temperature would be in the range of from 200°C to 260°C, most typically from 230°C to 250°C. For the production of staple fibres, a typical extrusion temperature would be in the range of from 230°C to 330°C, most typically from



280°C to 300°C.

The fibres produced in accordance with the invention may be produced from multimodal sPP/iPP blends having other additives to improve the mechanical processing or spinnability of the fibres. The fibres produced in accordance with the invention may be used to produce non-woven fabrics for use in filtration; in personal care products such as wipers, diapers, feminine hygiene products and incontinence products; in medical products such as wound dressings, surgical gowns, bandages and surgical drapes; in protective covers; in outdoor fabrics and in geotextiles. Non-woven fabrics made with the bimodal sPP/iPP fibres of the invention can be part of such products, or constitute entirely the products. As well as making non-woven fabrics, the fibres may also be employed to make a woven knitted fabric or mat. The non-woven fabrics produced from the fibres in accordance with the invention can be produced by several processes, such as air through blowing, melt blowing, spun bonding or bonded carded processes. The fibres of the invention may also be formed as a non-woven spunlace product which is formed without thermal bonding by fibres being entangled together to form a fabric by the application of a high pressure-fluid such as air or water.

The present invention will now be described in greater detail with reference to the following non-limiting example.

#### Example 1 and Comparative Example 1

In accordance with Example 1, a bimodal sPP having a dispersion index D of 3.9 and an MFI of 4.4g/10min was blended in an amount of 2wt% based on the weight of the blend with an isotactic polypropylene having the properties of Polymer 1 specified in Table 1. The blend was then subjected to spinning through a spinnerette to form fibres and the maximum spinning speed before

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breakage of the fibres was measured at two spinning temperatures. The results are shown in Table 2.

In accordance with Comparative Example 1, a monomodal sPP having a dispersion index D of 2.6 and an MFI of 4.5g/10min, *i.e.* an MFI substantially the same as that of the bimodal sPP of Example 1, was blended, also in an amount of 2wt% based on the weight of the blend, with the same isotactic polypropylene employed in Example 1. The resultant blend was similarly spun to form fibres at the same temperatures as for Example 1 and the results are also shown in Table 2.

It may be seen that for both spinning temperatures of 260°C and 280°C, the use of bimodal sPP substantially increases the maximum spinning speed before breakage of the fibres as compared to the use of the same amount of monomodal sPP blended with the same isotactic polypropylene.

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TABLE 1

		Bimodal sPP	iPP - Polymer 1	iPP Polymer 2
MI <sub>2</sub>	g/10min	3.6	25.9	14
Tm	°C	110 and 127	163.4	164.3
Mn	kDa	37426	35756	41983
Mw	kDa	160229	192383	256895
Mz	kDa	460875	755427	1173716
Mp	kDa	50516	96230	107648
D		4.3	5.4	6.1

TABLE 2

	Example 1	Comparative Example 1
Property	iPP-2wt% bimodal sPP	iPP-2wt% monomodal sPP
Maximum spinning speed at 260°C	3700	3300
Maximum spinning speed at 280°C	3100	2900

CLAIMS:

1. A polypropylene blend including from 0.3 to 50% by weight of a syndiotactic polypropylene having a multimodal molecular weight distribution and at least 50% by weight of an isotactic polypropylene.
2. A blend according to claim 1 wherein the multimodal sPP concentration in the sPP/iPP blend is from 0.5 to 15wt%.
3. A blend according to claim 2 wherein the multimodal sPP concentration in the sPP/iPP blend is from 1 to 10wt%.
4. A blend according to any foregoing claim wherein the iPP is a homopolymer, copolymer or terpolymer of isotactic polypropylene.
5. A blend according to any foregoing claim wherein the iPP has a dispersion index (D) of from 3.5 to 9, preferably 3.5 to 6.5.
6. A blend according to any foregoing claim wherein the iPP has a melting temperature in the range of from 159 to 169°C.
7. A blend according to any foregoing claim 4 to 6 wherein the iPP has an Mn of from 35,000 to 60,000 kDa.
8. A blend according to any foregoing claim wherein the iPP has a melt flow index (MFI) of from 1 to 90g/10mins.
9. A blend according to any foregoing claim wherein the multimodal sPP is a homopolymer or a random or block copolymer or a terpolymer.
10. A blend according to any foregoing claim wherein the

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multimodal sPP has a melting temperature of up to about 130°C.

11. A blend according to any foregoing claim wherein the multimodal sPP has an MFI of from 0.1 to 1000g/10min.

12. A blend according to claim 11 wherein the multimodal sPP has an MFI of from 1 to 60g/10min.

13. A blend according to any foregoing claim wherein the multimodal sPP has a Mn of from 35,000 to 40,000 kDa.

14. A blend according to any foregoing claim wherein the multimodal sPP has a dispersion index (D) of from 3 to 6.

15. A blend according to any foregoing claim wherein the multimodal sPP is bimodal.

16. A spun polypropylene fibre produced from the blend of any foregoing claim.

17. A fabric produced from the polypropylene fibre according to claim 16.

18. A product including a fabric according to claim 17, the product being selected from a filter, personal wipe, diaper, feminine hygiene product, incontinence product, wound dressing, bandage, surgical gown, surgical drape and protective cover.

19. Use, for increasing the maximum spinning speed when producing spun polypropylene fibres, of from 0.5 to 50wt% multimodal syndiotactic polypropylene in a blend with isotactic polypropylene.

# INTERNATIONAL SEARCH REPORT

International Application No

PC1/EP 00/02878

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 7 D01F6/46 C08L23/10

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 D01F C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 747 403 A (FINA TECHNOLOGY) 11 December 1996 (1996-12-11) example 1 table 1 figures 1,3	1-6,9, 10,13-15
X	US 5 648 428 A (REDDY BAIREDDY RAGHAVA ET AL) 15 July 1997 (1997-07-15) cited in the application column 6, line 33 - line 62 example 1 table 1 figures 1,3	1-6,9, 10,13-15
A	EP 0 634 505 A (HIMONT INC) 18 January 1995 (1995-01-18) cited in the application the whole document	1-19
	-/-	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/02878

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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Information on patent family members

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